

REMARKS

Claims 1-8 are currently pending. Claims 1 and 4 are currently amended. Claims 7 and 8 are newly added. Support for the amendments to claims 1 and 4 may be found in the specification as originally filed, for example, at page 11, line 20 to page 13, line 1. Support for new claims 7 and 8 may be found in the specification as originally filed, for example, in the definitions for formulae 1-3, page 11, lines 1-6 and page 34, lines 1-3.

I. The Rejection Under 35 U.S.C. §112

Claims 1-6 are rejected under 35 U.S.C. 112, first paragraph, as allegedly failing to comply with the enablement requirement.

Claims 1-6 are rejected under 35 U.S.C. 112, first paragraph, because the specification, while being enabling for particular disclosed derivative for each material claimed, allegedly does not reasonably provide enablement for all derivatives of such materials.

Claims 1-6 are rejected under 35 U.S.C. 112, second paragraph, as allegedly being indefinite.

For each of the enablement and indefinite rejections the Examiner is concerned with the scope and meaning of the term “derivative.”

First of all, Applicants respectfully submit that one skilled in the art would understand the term “derivative”, which is a well known term in the chemical arts and that one skilled in the art would be able to make and use such derivative compounds. However, to advance prosecution, Applicants have amended the claims to remove the term “derivative.” For example, “a vinylene carbonate derivative” has been rewritten to “a vinylene carbonate compound represented by

Chemical Formula 1,” with the Chemical Formula 1 defined. Likewise, “a cyclic sulfite derivative” has been rewritten to “a cyclic sulfite compound represented by Chemical Formula 2 or 3”. Phenylcycloalkane derivative and alkylbenzene derivative have been rewritten to phenylcycloalkane compound and alkylbenzene compound, respectively.

For the above reasons, it is respectfully submitted that Applicants’ claims are clear and definite and fully enabling to one skilled in the art and it is requested that the rejections under 35 U.S.C. §112 be reconsidered and withdrawn.

II. The Rejections Under 35 U.S.C. §103

Claims 1 and 3 are rejected under 35 U.S.C. 103(a) as allegedly being unpatentable over JP 2002-025611 A (JP '611) in view of JP 2002-298909 (JP '909), JP 2002-110229 (JP' 229) or JP 2002-050398 (JP '398).

Claim 2 is rejected under 35 U.S.C. 103(a) as allegedly being unpatentable over JP '611 in view of JP '909, JP '229 or JP '398, and further in view of U.S. Patent No. 6,818,351 (Sunagawa).

Claims 4 and 6 are rejected under 35 U.S.C. 103(a) as allegedly being unpatentable over JP '611 in view of JP '398 and either JP '909 or JP '229.

Claim 5 is rejected under 35 U.S.C. 103(a) as allegedly being unpatentable over JP '611 in view of JP '398 and either JP '909 or JP '229 and further in view of Sunagawa.

The Examiner’s basic position is that JP '611 discloses all of Applicants’ claimed elements except that JP '611 does not teach an electrolyte including a phenylcycloalkane

derivative or an alkylbenzene derivative having a quaternary carbon directly bonded to a benzene ring.

The Examiner cites JP '909 and JP '229 as disclosing adding tert-alkylbenzene derivatives to a lithium battery nonaqueous electrolyte.

The Examiner cites JP '398 as disclosing adding phenylcyclohexane to a lithium battery nonaqueous electrolyte.

As to claim 2, the Examiner cites Sunagawa as teaching the use of lithium cobalt compound oxides with bulk density of 3.3 g/cm^3 .

Applicants respectfully submit that the present invention is not anticipated by or obvious over the disclosures of the cited art and request that the Examiner reconsider and withdraw these rejections in view of the following remarks.

For the convenience of the Examiner, attached are Partial English language translations of JP'611, JP'909 and JP'398. A partial translation of JP'229 is not included. JP'229 and JP'909 have the same applicant and inventors and are almost the same in content of specification, although they differ in their claimed compound(s).

The present invention is for the purpose of solving the following problems:

It is impossible to inhibit the decomposition of the electrolyte solution when the cell is exposed to a high temperature (approximately 100°C) or when charge and discharge are repeated at around the upper limit (40 to 60°C) of normal use temperature of a cell;

There is a danger of solution leakage caused by an increased internal pressure resulting from solvent decomposition, when the cell is exposed to a high temperature (approximately

100°C). Furthermore, there is a problem of smoking or the like when the cell is heated abnormally by overcharge; and

An increase in the internal resistance of the cell poses the problem of high cycle deterioration.

See Applicants' specification, page 3, line 16-page 4, line 15.

The advantageous effects of the present invention are as follows:

Even when the cell temperature is high (105°C), there is no solution leakage and no unnecessary operation of the current-cutting device (see Applicants' specification at, for example, page 19, line 15-page 20, line 8, and Figs. 1-3); and

Degradation of the cycle characteristics is kept at low level even when the positive electrode active material density is increased (See Applicants' specification, Fig. 4).

JP'611 discloses a non-aqueous electrolyte secondary cell having vinylene carbonate and cyclic sulfite. However, JP '611 does not disclose a phenylcycloalkane compound having a quaternary carbon directly bonded to a benzene ring, or an alkylbenzene compound having a quaternary carbon directly bonded to a benzene ring. Further, JP '611 does not disclose "an explosion-proof valve mounted in the sealing structure, wherein the explosion-proof valve is capable of separating from the sealing plate to discontinue current when the internal cell pressure exceeds a predetermined value."

Since the teachings of JP '611 do not include explosion-proof valves, JP '611 is not for the purpose of solving the technical problem of "prevention of unnecessary operation of the current-cutting device".

Furthermore, the teachings of JP'611 are not for the purpose of solving the technical problem "prevention of solution leakage when the cell temperature is high (100°C or more)." In addition, JP '611 do not teach or disclose the purpose of solving the technical problem of "inhibition of degradation of cycle characteristics occurring when the bulk density of the active material is increased in order to increase cell capacity."

JP'909 and JP'229 both disclose adding a t-alkylbenzene derivative in the non-aqueous electrolyte of a lithium cell. However, neither of the references discloses "an explosion-proof valve mounted in the sealing structure such that the explosion-proof valve is capable of separating from the sealing plate to discontinue current when the internal cell pressure exceeds a predetermined value."

Further, neither of the references has a cyclic sulfite derivative.

Furthermore, the references do not teach or disclose the purpose of solving the technical problem of "prevention of solution leakage when the cell temperature is high (100°C or more)." In addition, the references do not teach or disclose the purpose of solving the technical problem of "inhibition of degradation of cycle characteristics occurring when the bulk density of the active material is increased in order to increase cell capacity."

Since the references do not have explosion-proof valves, they are not for the purpose of solving the technical challenge "prevention of unnecessary operation of the current-cutting device" does not exist in the references.

JP'398 discloses adding phenylcyclohexane in the non-aqueous electrolyte of a lithium cell. However, JP'398 does not have a cyclic sulfite derivative.

Further, the charging control system for detecting the cell temperature and cutting off the charging circuit is by a thermistor or a temperature fuse (see JP'398, paragraphs [0030] and [0046]). Thus, JP'398 does not have "an explosion-proof valve mounted in the sealing structure, wherein the explosion-proof valve is capable of separating from the sealing plate to discontinue current when the internal cell pressure exceeds a predetermined value."

As clearly seen from the above discussions, even if the teachings of JP'611, JP'909, JP'229, and JP'398 are combined, Applicants' claimed invention would not be obvious.

Further, the combination of JP'611, JP'909, JP'229, and/or JP'398 would not motivate or teach one of ordinary skill to prevent the unnecessary operation of the current-cutting device. The cited art are not for the purpose of solving disadvantages resulting from increasing the bulk density of the active material. Thus, one of ordinary skill would not find that selecting the above three substances recited in the references could solve the disadvantages. See also the unexpected results achieved by using the combination as claimed shown in the tables of Applicants' specification.

Sunagawa does not overcome the deficiencies in the primary references discussed above.

For the above reasons, it is respectfully submitted that the subject matter of claims 1-8 is neither taught by nor made obvious from the disclosures of JP '611 in view of JP 909, JP '229 or JP '398, or further in combination with Sunagawa, or the disclosures of JP '611 in view of JP '398 and either JP '909 or JP '229, or further in combination with Sunagawa, and it is requested that the rejections under 35 U.S.C. §103(a) be reconsidered and withdrawn.

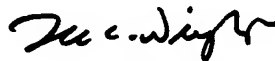
III. Conclusion

In view of the above, Applicants respectfully submit that their claimed invention is allowable and ask that the rejections under 35 U.S.C. §112 and the rejections under 35 U.S.C. §103 be reconsidered and withdrawn. Applicants respectfully submit that this case is in condition for allowance and allowance is respectfully solicited.

If any points remain at issue which the Examiner feels may be best resolved through a personal or telephone interview, the Examiner is kindly requested to contact the undersigned at the local exchange number listed below.

If this paper is not timely filed, Applicants respectfully petition for an appropriate extension of time. The fees for such an extension or any other fees that may be due with respect to this paper may be charged to Deposit Account No. 50-2866.

Respectfully submitted,
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Enclosures: Partial Translation of JP2002-25611;
Partial Translation of JP2002-298909; and
Partial Translation of JP2002-50398.

Partial Translation of JP2002-25611

[0008]

[PROBLEMS TO BE SOLVED BY THE INVENTION]

The present invention has been accomplished to solve these problems encountered in the prior art. Specifically, it is an object of the present invention to provide a nonaqueous electrolyte secondary cell with high-energy density that minimizes decomposition of the electrolyte of the nonaqueous electrolyte secondary cell and a change in the composition of the electrolyte, and that excels in storage characteristics and in cycle characteristics.

[0009]

[MEANS TO SOLVE THE PROBLEMS]

In order to solve the above problems, in the present invention, the specific surface area of the positive electrode active material is $0.25\text{--}0.75\text{ m}^2/\text{g}$, and the electrolyte contains ethylene-sulfite and vinylene carbonate. In particular, ethylene-sulfite is preferably contained at $0.5\text{--}2.0\text{ weight\%}$, and vinylene carbonate is preferably contained at $0.5\text{--}3.0\text{ weight\%}$. This has been found to provide a nonaqueous electrolyte secondary cell that excels in storage characteristics and in cycle characteristics.

[0015] (Example 1)

Fig. 1 shows a longitudinal section of the nonaqueous electrolyte secondary cell of the present invention.

[0016] The cell was prepared according to the following procedure. LiCoO_2 serving as the positive electrode active material was synthesized by mixing Li_2CO_3 and Co_3O_4 at a Co/Li mole ratio of $0.97\text{--}1.04$ which were then heated 900°C . It was possible to arbitrarily set the specific surface area of the positive electrode active material within the range $0.25\text{--}0.75\text{ m}^2/\text{g}$ by varying the Co/Li mole ratio. Then, the resulting product was classified into 100 mesh or lower, thus preparing the positive electrode active material.

[0021] The nonaqueous electrolyte was prepared as follows. A solvent in which 1 weight % of ethylene sulfite (ES) and 2 weight % of vinylene carbonate (VC) were added to a mixture of ethylene carbonate, methyl-ethyl carbonate, and dimethyl carbonate (1:1:1 volume ratio). Then in this solvent, LiPF_6 serving as solute was dissolved at 1 mole/litter.

[0022] (Comparative Example) A cell was prepared in the same manner as Example 1 except that the solvent of the electrolyte did not contain ethylene sulfite and vinylene carbonate.

[0023] (Example 2) Example 2 shows the results of optimization of ethylene sulfite and vinylene carbonate contained in the electrolyte by using a positive electrode active material with a specific surface area of 0.49 m²/g.

[0024] Cells were prepared in the same manner as Example 1 using electrolytes containing ethylene sulfite and vinylene carbonate at 0.2, 0.5, 1.0, 2.0, 3.0, and 5.0 weight %.

[0025] The cells of Examples 1 and 2 and Comparative Example were subjected to a discharging test at a discharging current of 1A and at 20°C. Also, the cells in a charged state were stored at 60°C for 20 days to compare the capacity ratios before and after storage. Furthermore, the cells were subjected to a cycle test at 45°C.

[0026] Fig. 2 shows the capacity ratios before and after storage of the cells of Examples 1 and 2 and Comparative Example, together with the discharging capacity retention rates after 100 cycles of 45°C. As long as the specific surface area of the positive electrode was 0.75 m²/g or lower, it was found that the cells containing both ethylene sulfite and vinylene carbonate had excellent storage characteristics and cycle characteristics as compared with those without ethylene sulfite and vinylene carbonate. It is considered that the improvement of the characteristics was because the contained ethylene sulfite and vinylene carbonate were decomposed on the surface of the positive electrode to form a proper covering film, which eliminated direct contact of the main solvent of the electrolyte and the electrode surface, thereby preventing decomposition of the electrolyte.

[0028] Table 1 shows the capacity retention rates before and after storage at 60°C of the cells prepared in Example 2. Table 2 shows the capacity retention rates after 100 cycles of 45°C along with the amount of the contained ethylene sulfite (ES) and vinylene carbonate (VC).

[0031] As seen from the Tables, the optimal range for vinylene carbonate is 0.5-3 weight %. It is also seen that the optimal range for ethylene sulfite is 0.5-2 weight %.

Partial Translation of JP2002-298909

[0003]

[PROBLEMS TO BE SOLVED BY THE INVENTION]

However, there is a need for secondary cells having more improved cell characteristics including cycle characteristics and electrical characteristics. For a lithium secondary cell using, as the positive electrode, LiCoO_2 , LiMn_2O_4 , or LiNiO_2 , for example, the solvent in the nonaqueous electrolyte is locally oxidative-decomposed during charging, and the decomposed matter prevents the cell from exhibiting the desired electrochemical reaction, resulting in degraded cell performance. Arguably this is caused by the electrochemical oxidation of the solvent at the interface of the positive electrode material and the nonaqueous electrolyte. For a lithium secondary cell using, as the negative electrode, a highly-crystallized carbon material such as natural graphite and artificial graphite, the solvent in the nonaqueous electrolyte is reductive-decomposed on the negative electrode surface during charging. Even EC, which is widely used as a nonaqueous electrolyte solvent, suffers reductive decomposition locally through repeated charging and discharging. This causes degradation of cell performance. Thus, the present situation is that cell characteristics including cycle characteristics and electrical characteristics are not satisfactory.

[0004] In order to solve the above problems associated with the nonaqueous electrolyte solvent for lithium secondary cells, it is an object of the present invention to provide such a nonaqueous electrolyte for a lithium secondary cell that can constitute a lithium secondary cell excellent in cell characteristics including electric capacity and charged-state storage characteristics. It is another object of the present invention to provide a lithium secondary cell utilizing the nonaqueous electrolyte.

[0024] [Preparation of Lithium Secondary Cell and Measurement of Cell Characteristics]

Eighty weight % of LiCoO_2 (positive electrode active material), 10 weight % of acetylene black (a conductive agent), and 10 weight % of polyvinylidene fluoride (a binder) were mixed. Added to this mixture was a 1-methyl-2-pyrrolidone solvent. Then, the resultant product was applied onto an aluminum foil, followed by drying, pressure-molding, and heat treatment, thus preparing a positive electrode. Ninety weight % of artificial graphite (negative electrode active material) and 10 weight % of

polyvinylidene fluoride (a binder) were mixed. Added to this mixture was a 1-methyl-2-pyrrolidone solvent. Then, the resultant product was applied onto a copper foil, followed by drying, pressure-molding, and heat treatment, thus preparing a negative electrode. Then, using a separator of a polypropylene microporous film, injection of the above-described nonaqueous electrolyte was carried out, thus preparing a coin cell (20 mm in diameter and 3.2 mm thick). This coin cell was charged at a constant current of 0.8 mA and a constant voltage to an end voltage of 4.2 V at room temperature (20°C) for five hours. The coin cell was then discharged a constant current of 0.8 mA to an end voltage of 2.7 V. The charging and discharging were repeated. The initial charging-discharging capacity was approximately equal to the case (Comparative Example 1) where 1M LiPF₆-EC/PC/DEC (volume ratio: 30/5/65) without a tert-alkylbenzene derivative contained therein was used as the nonaqueous electrolyte. The cell characteristics after 50 cycles were measured, and it was observed that the discharging capacity retention rate was 92.8% when assuming the initial charging-discharging capacity as 100%. It was also observed that the low-temperature characteristics were preferable as well. Table 1 shows the conditions for preparing the coin cell and the cell characteristics.

[0038]

Table 1

	Positive Electrode	Negative Electrode	Compound	Amount (wt%)	Composition of electrolyte (volume ratio)	The initial discharging capacity (relative value)	50 cycle discharging capacity retention rate (%)
Ex. 1	LiCoO ₂	Artificial graphite	Tert- pentylbenzene	2.0	1M LiPF ₆ EC/PC/DEC =30/5/65	1.03	92.8
Ex. 2	LiCoO ₂	Artificial graphite	Tert- pentylbenzene	5.0	1M LiPF ₆ EC/PC/DEC =30/5/65	1.02	91.5
Ex. 3	LiCoO ₂	Artificial graphite	Tert- pentylbenzene	0.5	1M LiPF ₆ EC/PC/DEC =30/5/65	1.01	90.3
Com. Ex. 1	LiCoO ₂	Artificial graphite		0	1M LiPF ₆ EC/PC/DEC =30/5/65	1.00	82.6
Ex. 4	LiCoO ₂	Artificial graphite	1-methyl-4- tert- pentylbenzene	2.0	1M LiPF ₆ EC/PC/DEC =30/5/65	1.02	92.1
Ex. 5	LiCoO ₂	Artificial graphite	(1,1-diethyl propyl)benzene	2.0	1M LiPF ₆ EC/PC/DEC =30/5/65	1.02	91.9
Ex. 6	LiNi _{0.8} Co _{0.2} O ₂	Natural graphite	Tert- pentylbenzene	2.0	1M LiPF ₆ EC/PC/DEC/DMC =30/5/30/35	1.02	92.8
Ex. 7	LiCoO ₂	Artificial graphite	Tert- pentylbenzene	2.0	1M LiPF ₆ EC/PC/MEC/DMC =30/5/50/15	1.15	91.1
Ex. 8	LiMn ₂ O ₄	Artificial graphite	Tert- pentylbenzene	2.0	1M LiBF ₄ EC/PC/DEC/DMC =30/5/30/35	0.99	92.6
Ex. 9	LiCoO ₂	Artificial graphite	4-fluoro-tert- pentylbenzene	3.0	1M LiPF ₆ EC/PC/DEC	1.02	92.7

					=30/5/65		
Com. Ex. 2	LiCoO ₂	Artificial graphite	toluene	3.0	1M LiPF ₆ EC/PC/DEC =30/5/65	0.98	81.3
Com. Ex. 3	LiCoO ₂	Artificial graphite	n-butyl benzene	3.0	1M LiPF ₆ EC/PC/DEC =30/5/65	0.97	79.7
Com. Ex. 4	LiCoO ₂	Artificial graphite	di-n-butyl phthalate	3.0	1M LiPF ₆ EC/PC/DEC =30/5/65	0.97	78.1
Com. Ex. 5	LiCoO ₂	Artificial graphite	4-fluoro- toluene	3.0	1M LiPF ₆ EC/PC/DEC =30/5/65	0.98	80.6

Partial Translation of JP2002-50398

[0013] The present invention is for solving these problems and provides an excellent cell that preferably maintains cycle characteristics and storage characteristics, which are essential characteristics for cells, and secures safety after overcharging.

[0030] The cutting-off of the circuit of charging following detection of an increase in the cell temperature is preferably by a positive characteristic thermistor (PTC) or a temperature fuse, and the positive characteristic thermistor (PTC) and the temperature fuse are most reliable when their operating temperature is within 60-120°C.

[0046] Furthermore, the charging control system of a non-aqueous electrolyte secondary cell comprises a positive characteristic thermistor (PTC) or a temperature fuse to function as to cut off the circuit of charging following detection of an increase in the cell temperature. This configuration provides a more reliable secondary cell system.

[0089] Next, 5 cell samples each from the cells 1-25 of the present invention and the comparative cells 26-29 were subjected to constant-voltage charging at a voltage of 4.2 V, a limited current of 500 mA, an environment temperature of 20°C for two hours. In this charged state, the discharging characteristics of the cell samples were measured at 1A. Then, the cell samples in the charged state were stored at 80°C for five days, and after the storage, the cell samples were subjected to charging and discharging under the same conditions. The capacity recovery rates (capacity after storage/capacity before storage $\times 100$ (%)) of the cell samples after the charging and discharging were obtained. The results are shown in Tables 1-6.

[0090] Twenty cell samples were prepared each from the above cells, and these charged cell samples were further subjected to overcharging at 1A in order to see whether there was abnormal heating of the cells. Tables 1-6 show the number of cells among the 20 cells which exhibited abnormal heating.

[0091]

Table 1

(Example 1)

No.	Additives	HOMO	LUMO	Recovery rates after storage	Overcharging test
1	hexaphenylbenzene	-9.442	0.0813	91.2	0/20
2	p-terphenyl	-9.3829	0.0474	93.7	0/20
3	1-phenylpiperazine	-8.8102	0.3045	94.8	0/20
4	1,2,3,4-tetrahydroisoquinoline	-9.1063	0.3747	92.8	0/20
5	phenylcyclohexane	-9.4288	0.3961	92.3	0/20
6	1,3,5-triphenyl benzene	-9.5698	0.1704	93.7	0/20
7	dodecahydrotriphenylene	-8.8282	0.5063	91.4	0/20
8	divinylbenzene	-9.0738	-0.1222	91.7	0/20
9	1,4-dicyclohexylbenzene	-9.2999	0.3508	91.7	0/20

[0096]

Table 6

(Comparative Examples 1 and 2)

No.	Additives	HOMO	LUMO	Recovery rates after storage	Overcharging test
26	.			93.9	8/20
27	biphenyl	-9.1449	-0.1419	78.4	0/20
28	thiophene	-9.5429	-0.1917	72.3	0/20
29	3-chlorothiophene	-9.2515	-0.4247	69.6	0/20